Supplementary Information for

Tracer-based investigation of organic aerosols in marine atmospheres from marginal seas of China to the northwest Pacific Ocean

Tianfeng Guo[1], Zhigang Guo[1], Juntao Wang[2], Jialiang Feng[3*], Huiwang Gao[2,4], Xiaohong Yao[2,4*]

[1] Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention, Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China;
[2] Lab of Marine Environmental Science and Ecology, Ministry of Education, Ocean University of China, Qingdao 266100, China
[3] School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China
[4]Pilot National Laboratory for Marine Science and Technology (Qingdao), Qingdao, China

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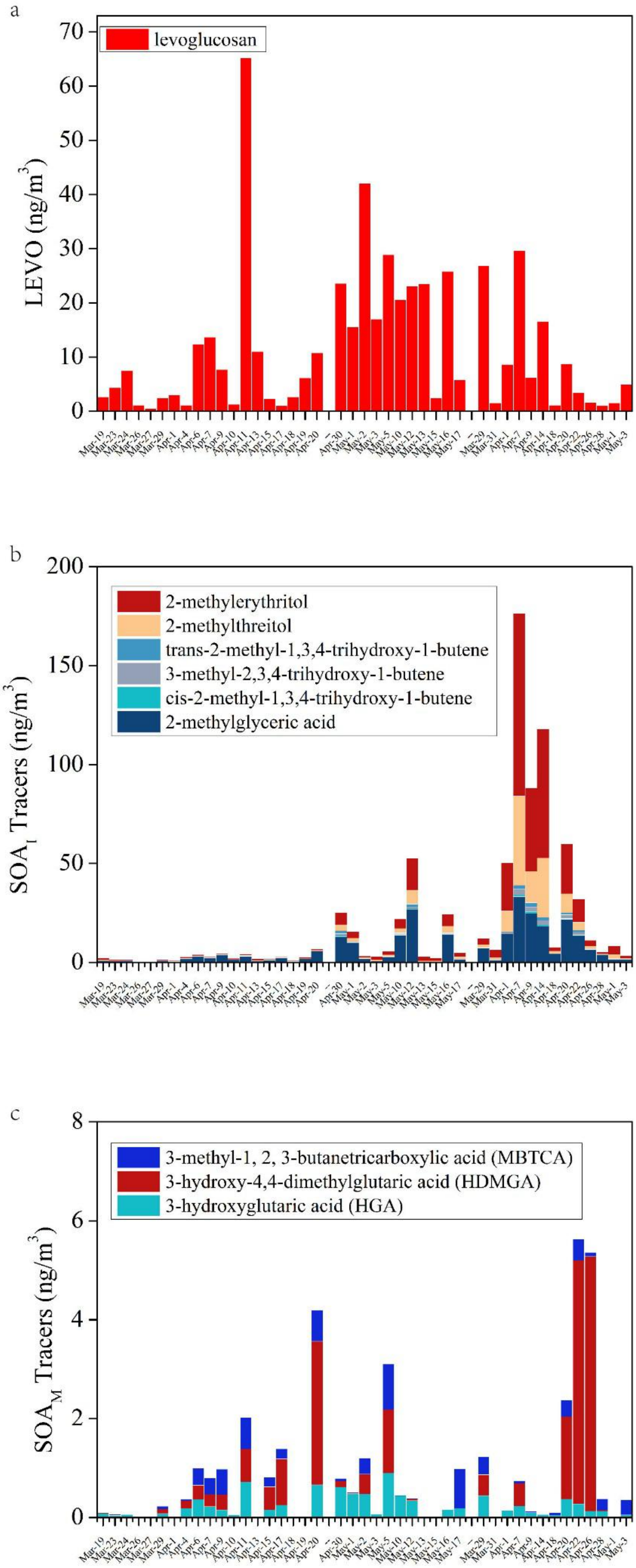
Section S1. Conversion between monoterpene SOA tracers analyzed in this study (HGA, HDMGA, MTBCA) and total monoterpene SOA tracers analyzed by Kleindienst et al. (2007). The SOA tracer method applies laboratory-determined ratios of the sum of specific SOA tracers to the total mass of SOA ($\frac{\sum SOA\ tracers}{total\ mass\ of SOA}$) to SOA tracers measured in the field (Kleindienst et al., 2007). Nine tracers, including pinonic acid, pinic acid, 2-hydroxy-4-isopropyladipic acid, 3-hydroxyglutanic acid (HGA), 3-hydroxy-4,4-dimethylglutaric acid (HDMGA), 3-isopropylpentanedioic acid, 3-acetylpentanedioic acid, 3-acetylhexanedioic acid and 3-(2-hydroxy-ethyl)-2-,2-dimethyl-cyclobutane-carboxylic acid, were used to estimate monoterpene SOA (Kleindienst et al., 2007). However, only three monoterpene SOA tracers were analyzed in this study, HGA, HDMGA and MBTCA, with two (HGA, HDMGA) common tracers used in both analyses. The correlation between the sum of these two common tracers and the sum of the nine tracers used in their study was analyzed. A strong correlation between the two common tracers (HGA+HDMGA) and the sum of all nine tracers indicated that they could be converted using a ratio of 0.32 (the slope, Fig. S4). Therefore, the total tracers $\Sigma_{tracers}$ and HGA+HDMGA values were converted using the formula (HGA+HDMGA) = 0.32* $\Sigma_{tracers}$. Thus, the f_{SOC} value for monoterpenes was scaled up by 3.1 based on laboratory observations, with the two tracers accounting for 2/9 of the total tracer level for monoterpenes (Kleindienst et al., 2007).

Section S2. Uncertainty of tracer-based SOC calculation

Both the quantification of SOA in ambient air and modeling of SOA remain challenging due to the variety of VOC sources and the complexity of SOA formation processes in the atmospheres of different environments (Hallquist et al., 2009). Under the assumptions that these organic tracers are stable in ambient air and that the tracer/OC conversion factors remain the same as those obtained from source samples or chamber simulations, the uncertainty of the SOA tracer method could be determined from analyses of the organic tracers and estimation of the appropriate conversion factors. The uncertainties in tracer analyses were less than 20% (Ding et al., 2008). The uncertainties of f_{SOC} was previously reported to be 25% for isoprene, 48% for monoterpenes, 22% for β-caryophyllene, and 33% for aromatics (Lewandowski et al., 2013). Considering these factors, the uncertainty in the estimated SOC values was calculated through error propagation. The relative standard deviations were 32% for SOC_I, 52% for SOC_M, 30% for SOC_C, and 39% for SOC_A.


a
levoglucosan
LEVO (ng/m3)
0
10
20
30
40
50
60
70
b
2-methylerythritol
2-methylthreitol
trans-2-methyl-1,3,4-trihydroxy-1-butene
3-methyl-2,3,4-trihydroxy-1-butene
cis-2-methyl-1,3,4-trihydroxy-1-butene
2-methylglyceric acid
SOAI Tracers (ng/m3)
0
50
100
150
200
c
3-methyl-1, 2, 3-butanetricarboxylic acid (MBTCA)
3-hydroxy-4,4-dimethylglutaric acid (HDMGA)
3-hydroxyglutaric acid (HGA)
SOAM Tracers (ng/m3)
0
2
4
6
8

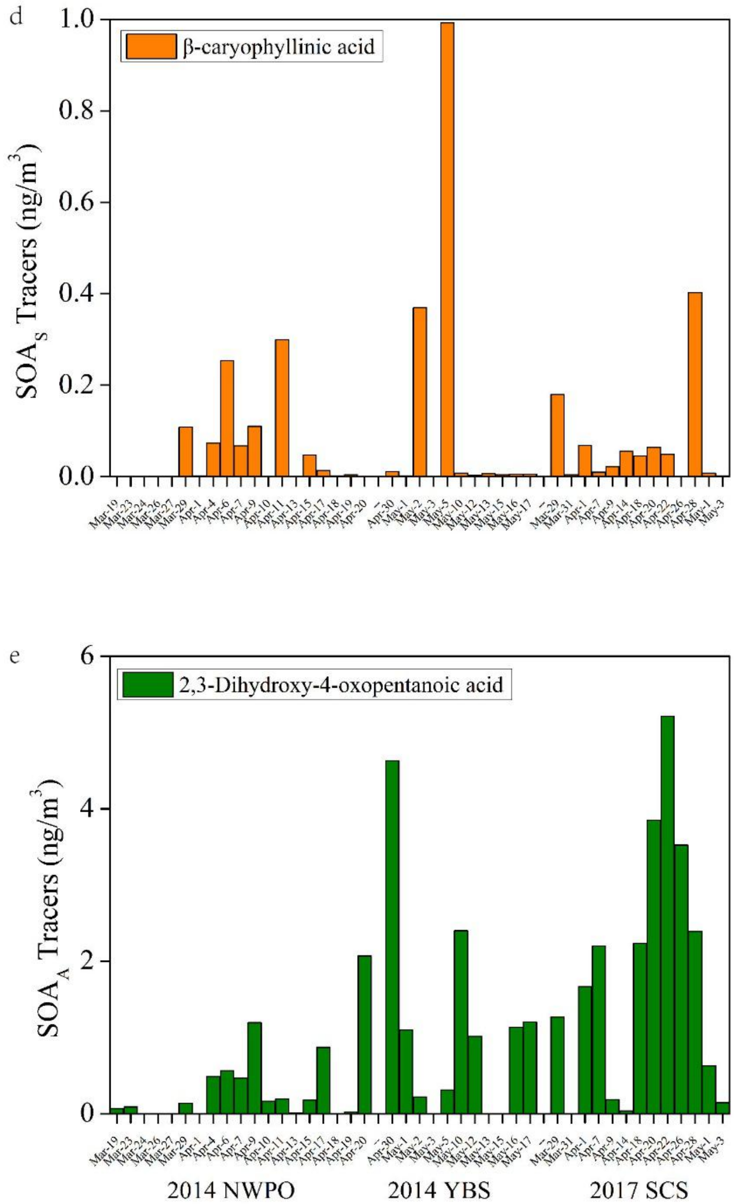


Figure S1. Concentration of primary and secondary organic tracers over the NWPO, YBS and SCS.

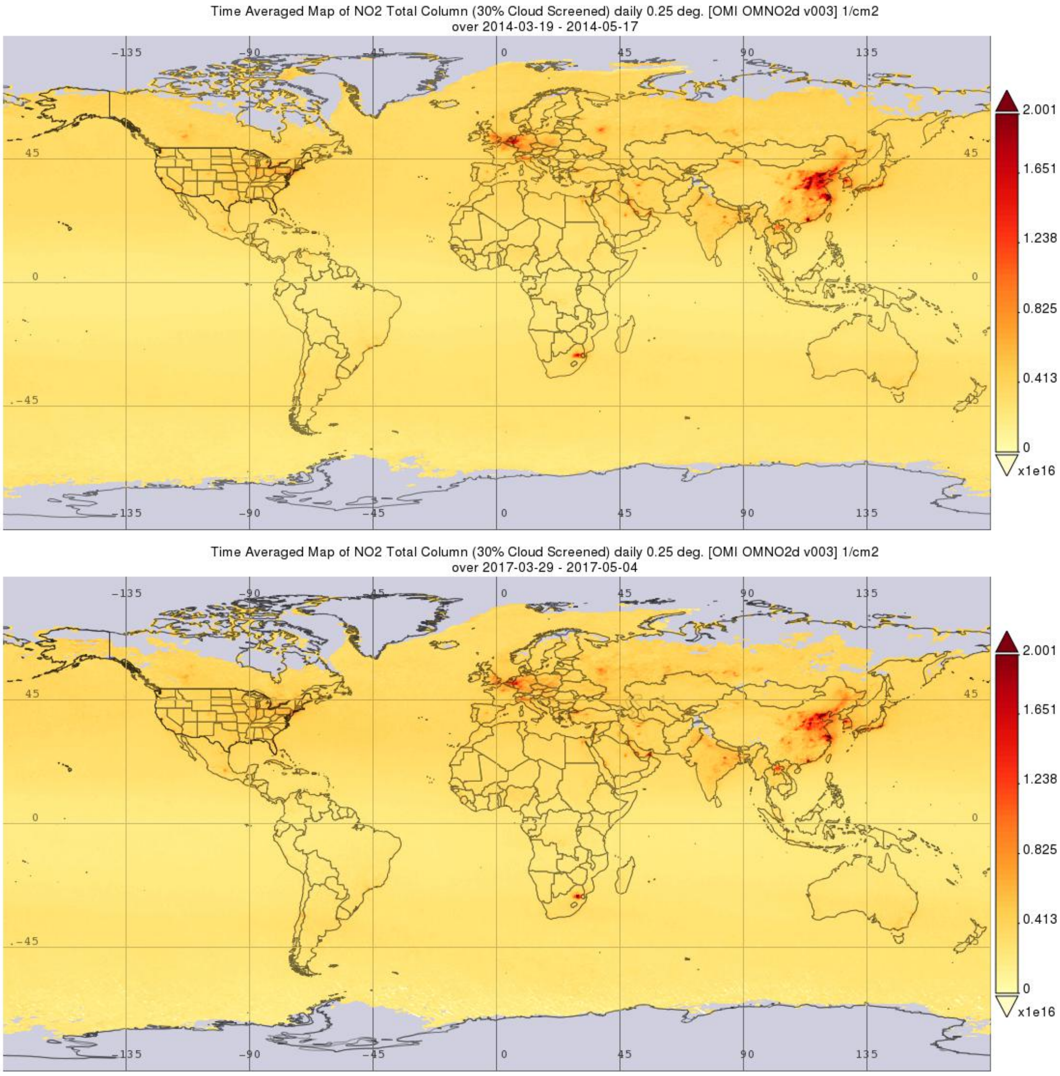


Figure S2. Average NO_2 distribution estimated globally using satellites in springtime during the sampling cruises in 2014 (up panel) and 2017 (below panel). Data are from https://giovanni.gsfc.nasa.gov/giovanni.

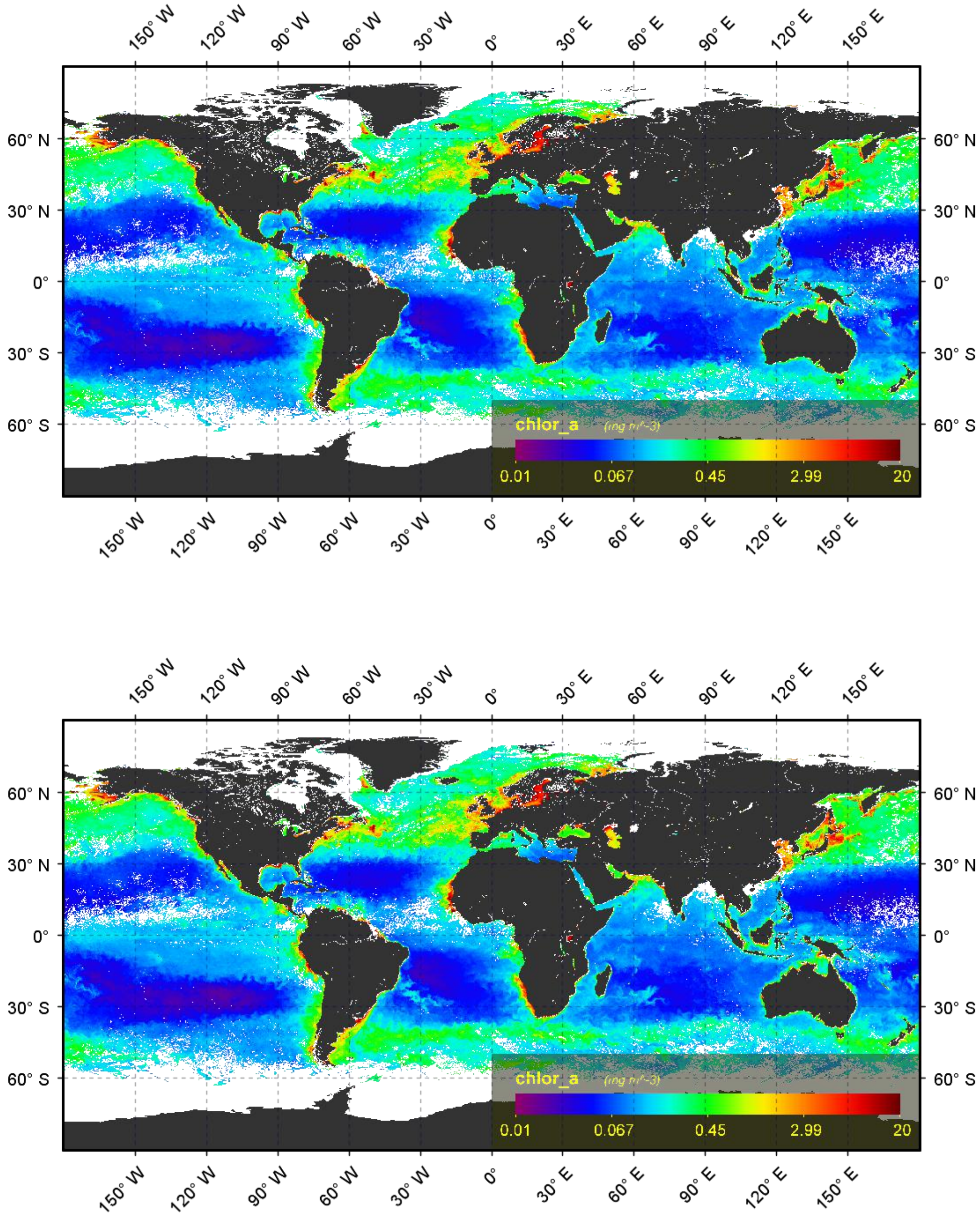


Figure S3. Monthly average marine chlorophyll-a distribution estimated globally using satellites in springtime (April) during the sampling cruises in 2014 (up panel) and 2017 (below panel). Data are from **https://oceancolor.gsfc.nasa.gov**.

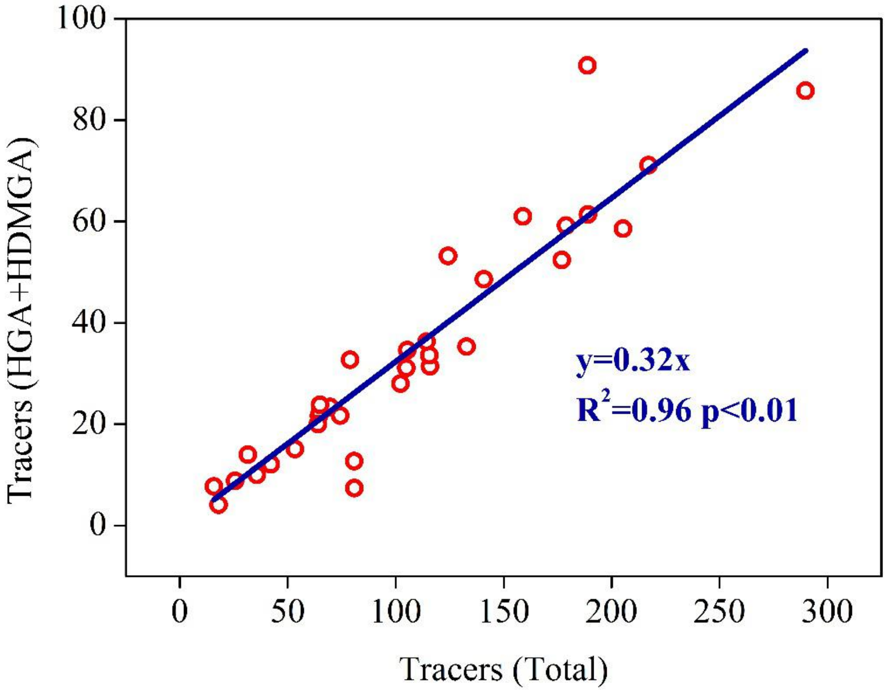


Figure S4. Linear correlation between two monoterpene SOA tracers (HGA+HDMGA) and nine monoterpene SOA tracers analyzed previously (Kleindienst *et al.*, 2007).

Table S1. Concentration of primary, secondary organic tracers (unit: ng/m^3); OC, EC (μg C/m^3) over the NWPO, YBS and SCS.

	gaactosan	mannosan	levoglucosan	2-methylglyceric acid	cis-2-methyl-1,3,4-trihydroxy-1-butene	3-methyl-2,3,4-trihydroxy-1-butene	trans-2-methyl-1,3,4-trihydroxy-1-butene	2-methylthreitol	2-methylerythritol	3-hydroxy glutaric acid	3-hydroxy-4,4-dimethylglutaric acid	3-methyl-1,2,3-butanetricarboxylic acid	β-caryophyllinic acid	2,3-Dihydroxy-4-oxopentanoic acid	OC	EC
2014 NWPO																
Mar-19	0.07	0.12	2.64	0.92	0.004	0.06	0.012	0.28	0.92	0.08	0.01	0.01	0.001	0.07	2.49	0.59
Mar-23	0.22	0.24	4.32	0.62	0.002	0.02	0.002	0.19	0.38	0.04	0.004	0.02	0.0002	0.09	1.49	0.31
Mar-24	0.24	0.30	7.49	0.91	nd	nd	nd	0.13	0.27	0.05	nd	nd	nd	nd	2.59	0.90
Mar-26	0.08	0.06	1.10	nd.	0.001	0.01	0.0004	0.06	0.17	0.001	0.001	nd	nd	nd	1.47	0.16
Mar-27	0.04	0.02	0.52	0.003	nd.	0.004	0.003	0.10	0.20	0.001	0.0001	nd	nd	0.001	1.27	0.35
Mar-29	0.15	0.17	2.44	0.88	0.002	0.01	0.003	0.14	0.32	0.08	0.09	0.06	0.11	0.14	2.62	1.76
Apr-1	0.20	0.20	2.95	0.21	0.001	0.02	0.001	0.24	0.49	0.00	0.00	0.00	0.002	0.00	1.21	0.13
Apr-4	0.09	0.05	1.13	1.76	0.001	0.03	0.003	0.34	0.69	0.19	0.15	0.03	0.07	0.49	0.91	0.23
Apr-6	0.61	0.64	12.38	2.98	0.002	0.02	0.004	0.29	0.59	0.37	0.28	0.34	0.25	0.56	1.16	0.13
Apr-7	0.68	0.70	13.61	2.35	0.004	0.02	0.003	0.20	0.42	0.22	0.25	0.33	0.07	0.47	2.15	0.30
Apr-9	0.19	0.26	7.65	3.72	nd	nd	nd	0.20	0.39	0.15	0.31	0.52	0.11	1.19	3.88	0.52
Apr-10	0.07	0.08	1.30	1.35	0.003	0.01	0.001	0.17	0.36	0.05	0.00	0.01	nd	0.17	1.23	0.24
Apr-11	3.60	3.64	65.18	3.07	0.009	0.03	0.011	0.32	0.67	0.72	0.66	0.64	0.30	0.19	4.25	0.55
Apr-13	0.88	0.86	11.02	0.61	0.005	0.05	0.002	0.37	0.76	nd	nd	nd	nd	0.01	3.59	0.57
Apr-15	0.12	0.15	2.27	1.11	0.002	0.02	0.002	0.24	0.50	0.16	0.46	0.20	0.05	0.18	1.80	0.16

Apr-17	0.07	0.08	1.02	2.31	0.003	0.01	0.001	0.15	0.33	0.25	0.93	0.20	0.01	0.87	1.09	0.12
Apr-18	0.08	0.14	2.68	0.20	nd	0.01	nd	0.19	0.42	nd	nd	nd	0.002	0.003	3.05	0.44
Apr-19	0.28	0.39	6.15	1.75	0.003	0.02	0.005	0.24	0.51	0.02	nd	0.004	0.004	0.02	8.88	3.59
Apr-20	0.40	0.63	10.76	5.66	0.014	0.05	0.015	0.31	0.65	0.66	2.91	0.63	nd.	2.07	6.65	0.74
2014 YBS																
Apr-30	0.86	1.70	23.53	13.03	0.62	0.81	1.58	2.88	6.32	0.62	0.12	0.05	0.01	4.63	8.95	2.26
May-1	0.98	1.90	15.53	9.91	0.09	0.32	0.26	1.67	3.30	0.48	0.01	0.01	0.002	1.10	8.63	1.19
May-2	3.16	5.98	42.03	1.90	0.01	0.01	0.03	0.45	0.86	0.48	0.40	0.33	0.37	0.22	10.42	1.99
May-3	2.54	3.88	16.96	0.52	0.01	0.02	0.01	0.72	1.63	0.07	nd	nd	nd	nd	7.46	1.04
May-5	3.20	4.48	28.86	2.79	0.03	0.10	0.05	0.91	1.69	0.90	1.28	0.92	0.99	0.31	9.84	2.56
May-10	1.23	2.16	20.55	13.80	0.22	0.46	0.48	2.23	4.79	0.44	nd	0.01	0.01	2.40	9.45	1.98
May-12	1.02	1.40	23.05	26.89	0.35	0.90	1.29	6.99	16.15	0.34	0.04	0.01	0.003	1.01	12.15	4.05
May-13	2.13	1.55	23.46	0.01	0.01	0.06	0.02	0.68	2.08	0.01	nd	0.0002	0.01	0.001	10.31	2.57
May-15	0.69	0.06	2.50	0.04	0.005	0.03	0.01	0.67	1.59	0.00	nd	0.001	0.004	0.002	16.29	6.36
May-16	1.14	1.62	25.77	14.00	0.04	0.40	0.54	3.32	5.97	0.16	nd	0.002	0.01	1.14	8.59	2.32
May-17	0.24	0.32	5.84	1.44	0.05	0.09	0.08	1.18	1.94	0.19	nd	0.80	0.01	1.20	8.53	1.87
2017 SCS																
Mar-29	nd	nd	26.79	7.17	0.05	0.14	0.09	1.56	3.17	0.44	0.42	0.36	0.18	1.27		
Mar-31	nd	nd	1.49	1.05	0.06	0.19	0.15	0.99	4.06	0.02	nd	nd	0.004	0.00		
Apr-1	nd	nd	8.60	14.32	0.07	0.89	0.27	10.35	24.32	0.14	nd	0.01	0.07	1.67		
Apr-7	nd	nd	29.61	33.15	0.83	2.98	2.02	44.99	92.48	0.23	0.46	0.05	0.01	2.20		

Apr-9	nd	nd	6.26	24.67	1.04	2.35	2.10	15.75	42.23	0.10	nd	0.02	0.02	0.18		
Apr-14	nd	nd	16.54	18.15	0.65	2.37	1.64	29.93	65.23	0.06	nd	nd	0.06	0.04		
Apr-18	nd	nd	1.06	4.35	0.03	0.09	0.08	0.98	2.04	0.04	nd	0.05	0.05	2.24		
Apr-20	nd	nd	8.71	21.74	0.47	1.91	0.97	9.68	25.01	0.37	1.67	0.32	0.06	3.85		
Apr-22	nd	nd	3.43	13.37	0.36	1.62	0.90	4.04	11.66	0.28	4.92	0.44	0.05	5.21		
Apr-26	nd	nd	1.61	6.54	0.09	0.33	0.16	1.18	2.91	0.13	5.15	0.08	nd	3.52		
Apr-28	nd	nd	1.00	3.83	0.02	0.07	0.04	0.38	0.85	0.13	0.02	0.23	0.40	2.39		
May-1	nd	nd	1.52	1.65	0.03	0.09	0.08	2.21	4.22	0.02	nd	nd	0.01	0.63		
May-3	nd	nd	4.95	1.54	0.03	0.09	0.08	0.45	1.32	0.05	nd	0.31	0.002	0.15		

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